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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

RE:

Patent Application Serial No. 09/485 034
(derived from International Application
PCT/GB99/01456 dated May 26, 1999)

of Robert John Blythe

Filed: February 2, 2000

For: CONSTRUCTION MATERIALS

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SEP 3 - 2002

TC 1700

DECLARATION

I, Robert John Blythe, of 53 Fairlawns, Yardley, Birmingham B26 2DT, England, declare as follows:

1. I hold the degree of Doctor of Philosophy from the Department of Transportation of the University of Birmingham, England.

I am an Associate of the National College of Rubber Technology.

I have given papers to professional bodies such as the American Society for the Testing of Materials, the International Rubber Conference and the Plastics and Rubber Institute. One such paper is published in Proceedings of the International Rubber Conference 1986, Volume 1 "Treadwear and Treadwear Mechanisms" p.p. 230 - 235. Published by Swedish Institution of Rubber Technology 1986.

I have been closely involved in the rubber and related

industries since 1970. A summary of my qualifications and experience is given in the following table:

Dates From	Dates To	Employer or Educational Body	Title	Relevant experience or qualification obtained
1970	1971	Dunlop (NCRT)	Student	Grad. IRI ANCRT
1971	1976	Dunlop	Rubber Compounder	Developing compounds for use in tyres. SBR included in range of polymers *
1976	1981	Dunlop	Senior Compounder	As above but responsible for a group of compounders Involved with a joint project between Dunlop Rubber Limited and Shell Internationale Research Maatschappij BV to develop new fuel-efficient tyre tread polymers (SBR) *
1981	1984	Dunlop + (University of Birmingham)	Student P/T	Research work leading to the Ph.D degree *
1984	1989	Dunlop	Manager Material Research	Research into new chemicals suitable for use in tyres including SBR. Development compounding for specialised use and passenger tyres SBR *
1989	1993	En-tous-Cas Limited	Development Manager	Developing new sports and leisure surfaces using granulated rubber, (SBR, NR, EPDM)
1993	1996	Leigh Environmental	Sales Manager	Technical, marketing and sales of recycled rubber granules
1996	1998	Rushden Granulating Limited	Technical Sales Manager	As above but including coloured EPDM. Developed SBS compound, variants. Marketing in USA.

1998	2000	Rushden Playsafe Limited	Technical Director	All technical functions including ongoing development of SBS compounds.
2000 to present date	-	Yardley Elastomeric Services	Senior Consultant	Working with rubber compounder to continue development of SBS compounds; co-operation with playground installers and users (granular SBR, NR, EPDM)

* Includes work on vulcanisation

Abbreviations used in the table have the following meanings:

SBR - styrene-butadiene copolymers
SBS - styrene-butadiene-styrene thermoplastic elastomers
NR - natural rubber
EPDM - ethylene-propylene-diene monomers

I am the inventor of numerous patents and patent applications relating to the use of elastomeric materials

Examples of those patents are:

<u>Patent No.</u>	<u>Subject</u>
US 4 845 154	Process for the production of a elastomeric copolymer of an aromatic vinyl compound and a conjugated diene
US 4 350 621	Elastomeric compositions
US 4 373 069	Elastomeric compositions
US 5 011 876	Pneumatic tires
US 5 115 021	Pneumatic tires
US 4 948 849	Process for making copolymers of aromatic vinyl compounds and conjugated diolefins
GB 2 236 109	Ground surface materials; sports surfaces

2. I am the inventor of Patent application Serial No. 09/485 034 ("the Application").

3. I have examined:

The patent specification of the Application;

the Official Action dated July 3, 2001;

the Final Rejection dated April 23, 2002; and

the citations in the names of Bowers, Thelen and Mitsui Toatsu referred to in the Final Rejection.

General

4. A major matter raised in the Final Rejection is the respective natures of thermoplastic elastomers (TPEs) and elastomers referred to as "SBR".

There is now shown to me an Exhibit marked "RJB 1" that is a copy of pages 15 to 20 of volume 9 of Encyclopaedia of Chemical Technology (Kirk-Othmer) which describes the nature of TPEs. In particular, page 15 summarises the principal distinguishing feature of TPEs: that they can exist as a processible melt and as a rubber-like solid, and that transition from the processible melt to the rubber-like solid takes place on cooling and is rapid and reversible. Distinction is drawn between TPEs and conventional rubber which must be cross-linked (i.e. vulcanised) to give useful properties and that vulcanisation, being a thermosetting process, is slow and irreversible and takes place upon heating.

Further, there is also shown to me an Exhibit marked "RJB2" that is a copy of page 400 of the Concise Encyclopaedia of Chemical Technology by Kirk-Othmer where it is stated, in the first paragraph of the left hand column:

"All types of SBR use compounding recipes, as do other unsaturated hydrocarbon polymers, that share the common ingredients of sulphur or extenders".

The reference in that quoted passage to the use of sulphur is to the use of sulphur as a vulcanisation agent. In my experience, sulphur is never used as a vulcanisation agent for polymers of the TPE type, nor is any other cross-linking agent.

Reference is now made to the three citations in turn.

5. International Patent Publication WO 92/19669 (Bowers)

Bowers is concerned with a method of producing a porous composite in which a particulate material is pre-wetted with a liquid curing agent to form a stable intermediate and, subsequently, the intermediate is treated with a liquid polymer or pre-polymer to form the desired composite. The resulting composite is said to be useful as an impact-absorbing surface or substrate for sports or play areas.

Nowhere in Bowers is there any disclosure or suggestion of the use of a TPE, as the particulate material or otherwise. It is clear from the disclosure as a whole that the nature of the particulate material is not critical; the invention is concerned rather with the provision of a stable, readily transportable intermediate which can be converted, at the intended site, relatively simply without the need for the use of expensive equipment or highly trained operators.

I note that in the description portion of the Bowers specification there is very little emphasis on, or detail of, the particulate material. However, at page 6, lines 12 to 15 it is stated: "the particulate material may comprise cork crumb, gravel, chipped bark or any other material suited to the intended application". Those specified materials are all cheap substances and all, with the exception of gravel, readily available in industry as waste products.

In the official action dated 23 April 2002 the Examiner has stated:

"Bowers teaches a polymeric material of the type contemplated by applicant, wherein said polymeric material can be used as a construction material in the formation of a (warring, sic) wearing course, said material comprises an agglomerate of a TPE as a block copolymer of the styrene type and a polyurethane binder which substantially filled the interstices between the granules, wherein the particle size is within applicant's claimed range".

In reference to the "polymeric material" I assume that the Examiner refers to the particulate material. However, my careful reading of Bowers has failed to identify any disclosure therein of the use as the particulate material of "a thermoplastic elastomer" or "a block copolymer of the styrene type". The only disclosures that I have been able to find relating to the use of a polymer as the particulate material are as follows:

Rubber crumb - Page 3, lines 10 to 11
Page 4, lines 7 to 10

Natural or synthetic rubber crumb -
Page 3, lines 10 to 11

Claim 8

None of those disclosures refers in any way to a "thermoplastic elastomer" or to "a block copolymer of the styrene type".

SBR waste rubber granules -

"SBR" is a commonly used abbreviation for synthetic rubbers obtained by the random co-polymerisation of styrene and butadiene by an emulsion polymerisation process; they are not block copolymers and, in particular, are not thermoplastic elastomers.

The general tenor of Bowers is that there should be used as the particulate material a cheap material such as that obtainable as a waste product in industry. Thermoplastic elastomers are not cheap materials. In general, their normal selling price is substantially greater than the normal selling price of a styrene-butadiene random copolymer sold under the designation "SBR".

It is to be noted that in the disclosure at page 3, lines 10 to 11 and at claim 8 natural rubber crumb is equated with synthetic rubber crumb. Natural rubber is neither a thermoplastic elastomer nor a block copolymer. The disclosure of "natural or synthetic rubber crumb" is a teaching to use as the particulate material granulated scrap vulcanised natural or synthetic rubber such as is readily obtainable from scrap motor tyres.

The Examiner has referred at page 3, lines 4 and 5 of the official action to Claims 7, 8 and 9 of application 09/485 034. It is to be noted that each of those claims is concerned with the use as TPE of a block copolymer and is thereby distinguished, for the reasons given above, from the rubber materials disclosed by Bowers for use as the particulate material.

6. US Patent 4 564 310 (Thelen)

Thelen is concerned with a paving system comprising four layers numbered (in Claim 1 and in the Abstract) as numbers 1 to 4. Layers 2, 3 and 4 comprise, respectively, scrap vulcanised rubber particles, scrap vulcanised rubber fibres and further scrap vulcanised rubber particles. None of those three scrap rubber materials can be a TPE for two reasons: first, as stated above, TPEs are not used in a vulcanised form. Secondly, to convert vulcanised rubber into a particulate form it is necessary to subject the vulcanised rubber to mechanical shearing which inevitably gives rise to a substantial increase of the temperature of the vulcanised rubber. For that reason TPEs are conventionally not reduced to particulate form by a mechanical shearing process as such a process could result in the TPE becoming molten and unworkable. Moreover, re-use of scrap TPE is much better effected by blending in the molten state.

I note that at column 3, lines 13 et seq Thelen states: "The core composition of our invention consists of a lattice, or matrix of vulcanised rubber fibres buffed or abraided from used tyres. . . ." The latter disclosures is exemplified by the Examples each of which discloses that the vulcanised rubber particles used had been abraided from vulcanised rubber tyres.

7. Japanese Patent 54048927 (Mitsui Toatsu)

Mitsui Toatsu is concerned with elastic playing surfaces comprising a base coated successively with a coarse granular layer, a fine granular rubber sealing layer and a synthetic resin surface layer. By definition. the

"granular" rubber layers, have been produced from a rubber material by a process of granulation. Although the abstract cited by the Examiner refers to the granular material being a "styrene-butadiene rubber", that rubber cannot be a TPE because, for the reasons stated above with reference to Thelen, a TPE is conventionally not subjected to a granulation process.

The undersigned declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that wilful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such wilful false statements may jeopardise the validity of the application or any patent issuing thereon.

Date:

8/8/02

Robert J Blythe

Robert John Blythe

R. E. Parr

Exhibit RJB 2

400 ELASTOMERS, SYNTHETIC

R. E. Parr Chartered Patent Agent

All types of SBR use compounding recipes, as do other unsaturated hydrocarbon polymers that share the common ingredients of sulfur, accelerators, antioxidants, antiozonants, activators, fillers, and softeners or extenders. SBR requires less sulfur than natural rubber for curing. The usual range is 1.5-2.0 phr of sulfur; however, this range should be based on the rubber hydrocarbon only for oil-extended SBR. Because of their lower unsaturation, all styrene-butadiene rubbers are slower curing than natural rubber and require more acceleration. Processing SBR compounds is similar to that of natural and polybutadiene rubbers. The ingredients are mixed in internal mixers or on mills and may be extruded, calendered, molded and cured in conventional equipment.

Uses

About 65% of all SBR elastomer produced in the United States is used in the manufacture of passenger-car tires. Two expanding markets for SBR are adhesives (qv) and chewing gum. A wide variety of SBRs is available for adhesive applications, and several of the crumb forms were designed specifically for the adhesives industry.

The block styrenic copolymers are intended for applications in adhesives, caulks, sealants, coatings, food packaging, toys, tubing, sheeting, molding equipment, belting, shoe soles and heels, and miscellaneous uses.

R. E. HAUER

The Goodyear Tire & Rubber Company

Table 2. Typical Properties of Segmented Polyether Esters

40T hard segment, %	33	58	76
polymer melt temperature (by dec), °C	176	202	212
specific gravity	1.16	1.20	1.22
tensile strength, MPa (psi)	39.3 (5700)	44.1 (6400)	47.5 (6900)
elongation at break, %	810	760	510
flexural modulus, MPa (psi)	44.8 (6500)	206 (30,000)	496 (72,000)
oil swell (ASTM NO. 3 oil, 7 days at 100°C), % vol increase	22.0	12.2	6.6

* Differential scanning calorimetry.

The SBS elastomers may be processed by a wide variety of techniques including solution processing, extrusion, calendering, injection molding, blow molding, and vacuum forming. Standard rubber and plastics equipment is useful for processing the elastomers.

Uses for the thermoplastic elastomers fall into two main sectors: primary raw materials for rubber products without vulcanization, and modifiers to upgrade the qualities of the rubbers and plastics. The largest markets for the styrene-diene block copolymers are footwear, adhesives (qv), and mechanical goods.

Thermoplastic Urethane Elastomers

Thermoplastic polyurethane (TPU) elastomers are a special class of urethanes that can be processed as plastics and as cements for a wide range of applications (see Urethane polymers). Generally, polyester-based materials are selected for high strength, tear, chemical and heat resistance, and polyether-based materials are selected for low temperature flexibility, high humidity conditions, and resistance to attack by fungi and bacteria.

Since urethane elastoplastics incorporate exceptional resistance to abrasion, fuel and oils, and have high tensile, tear, and load-bearing properties, and are available in a broad durometer range, they are candidates for demanding applications in such areas as automotive, sporting, general mechanical goods, fabric coatings, and biomedical applications such as intra-aortic balloons (see Prosthetic and biomedical devices).

Thermoplastic Copolyester-Ether Elastomers

Segmented copolyester-ethers represent a novel family of commercial thermoplastic elastomers derived from terephthalic acid (T), polytetramethylene ether glycol (PTMEG), and 1,4-butanediol. They offer an unusual combination of easy processing and high performance under environmental extremes (see Polyesters; Polyethers). The polyester-ether copolymers are prepared by titanate ester (tetrabutyl titanate)-catalyzed melt transesterification of a mixture of dimethyl terephthalate, polyether glycol, and excess 1,4-butanediol. Some typical physical properties are listed in Table 2.

The thermoplastic copolyester-ether elastomers commercialized as Hytrel by DuPont can be processed by injection, blow, compression, transfer, or rotational molding. Some of the many uses of these elastomers include as a replacement for cured rubber and rubber-metal parts with a one-component elastomer unit.

A. F. FINELLI
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D. A. CHUNG
The Goodyear Tire and Rubber Co.

THERMOPLASTIC ELASTOMERS

Thermoplastic resins are polymeric structures that soften or melt at elevated temperatures, allowing them to be processed into fabricated products that, when cooled, recover the physical and chemical properties of the original resin. Of the three classes of thermoplastic elastomers to be discussed, the styrene-diene block copolymers are the largest volume (> 50,000 metric tons), the thermoplastic polyurethanes are next (> 15,000 t), and copolyester ethers, the newest entry, are now > 2000 t.

Styrene-Diene Thermoplastic Block Copolymers

Preparation of styrene-diene block copolymers is achieved by forming a living polymer, a term coined to describe the product of a polymerization that has no termination or chain-transfer reactions. Shown in Table 1 is a comparison of mechanical properties of SBS block copolymers with vulcanized SBR and natural rubber illustrating the range inherent in the thermoplastic elastomers.

The chemical characteristics of the copolymers are determined by the nature of the components. Alteration of the chemical characteristics is achieved by altering one or more of the blocks.

The styrene-diene thermoplastic elastomers have excellent resistance to water, acids, and bases. Resistance to hydrocarbons, solvents, and oils is poor. The thermoplastic nature limits their utility to temperatures below 65°C depending on the stress. Elastic recovery, compression set, and creep properties are usually inferior to the chemically cross-linked elastomers.

Table 1. Typical Properties of ABA Thermoplastic Elastomers and Conventional Rubbers

	Kraton 1101 ^a	Kraton 1107 ^b	Natural rubber	SBR rubber
styrene, %	30	14		
tensile strength, MPa ^c	31.8	21.4	20.8	14.5
modulus at 300% ext, MPa ^c	2.8	0.7	3.5	2.1
elongation at break, %	880	1300	600	800
hardness, Shore A	71	37	65	46
specific gravity	0.94	0.92		

^a SIS (styrene-butadiene-styrene).

^b SIS (styrene-isoprene-styrene).

^c To convert MPa to psi, multiply by 145.

A. Nozay and J. E. McGrath, *Block Copolymers: Overview and Critical Survey*, Academic Press, Inc., New York, 1976.

U.S. Pat. 3,285,766 (Aug. 9, 1965), G. Holden and R. Milkovich (to Shell Oil Co.).

U.S. Pat. 2,871,218 (Jan. 27, 1959), C. S. Schollenberger (to B. F. Goodrich Co.).

U.S. Pat. 3,651,014 (Mar. 21, 1972), W. K. Wittepe (to E. I. du Pont de Nemours and Co., Inc.).

78. *Goodyear-Beaumont Chemical Plant Booklet*, Goodyear Chemical Division, The Goodyear Tire & Rubber Co., Akron, Ohio, 1992.
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86. B. Davis, *Rubber & Plastics News* 21(27), 6 (July 6, 1992).
87. Technical data, The Goodyear Tire & Rubber Co., Akron, Ohio, Aug. 1, 1992.
88. E. L. Puskas, Co., Akron, Ohio, Sales Agents and Distributors for Kuraray Co., Ltd. (Japan), *Trans-Polyisoprene TP-301*, Dec., 1991.
89. B. F. Greek, *Chem. Eng. News* 69(19), 37 (May 13, 1991).
90. E. J. Buckler, *Can. J. Chem. Eng.* 62(1), 3 (1984).
91. H. Takino and co-workers, *Kautsch. Gummi Kunstst.* 43(9), 761 (1990).

MICHAEL SENYEK

The Goodyear Tire & Rubber Co.

ELASTOMERS, SYNTHETIC-STYRENE-BUTADIENE RUBBER. See STYRENE-BUTADIENE RUBBER.

THERMOPLASTIC ELASTOMERS

Thermoplastic elastomers (TPE) were introduced in the 1960s. They have shown rapid growth since then and have been the subject of many conferences, symposia, etc. In particular, the developments in this field have been covered fairly recently in two books: one dealing primarily with the scientific aspects of these polymers (1) and the other dealing primarily with their applications (2). Thermoplastic elastomers have many of the physical properties of rubbers, ie, softness, flexibility, and resilience; but in contrast to conventional rubbers, they are processed as thermoplastics. Rubbers must be cross-linked to give useful properties. In the terminology of the plastics industry, vulcanization is a thermosetting process. Like other thermosetting processes, it is slow and irreversible and takes place upon heating. With thermoplastic elastomers, on the other hand, the transition from a processible melt to a solid, rubberlike object is rapid and reversible and takes place upon cooling. Thermoplastic elastomers can be processed using conventional plastics techniques, such as injection molding and extrusion; scrap is usually recycled.